

1 **Progress on Understanding Spatial and Temporal Variability of PM<sub>2.5</sub> and its**  
2 **Components in the Detroit Exposure and Aerosol Research Study (DEARS)**

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42 Spatial and temporal relationship of central site measures to indoor, outdoor, and personal  
43 exposure of the components of ambient  $PM_{2.5}$  (<http://www.epa.gov/DEARS/>).

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53 **1.0 Abstract**

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55 The Detroit Exposure and Aerosol Research Study (DEARS) measured personal exposures,  
56 ambient, residential indoor and residential outdoor concentrations of select PM<sub>2.5</sub> aerosol  
57 components (SO<sub>4</sub>, NO<sub>3</sub>, Fe, Si, Ca, K, Mn, Pb, Zn, EC and OC) over a three year period (2004-  
58 2007). These events represented approximately 190 calendar days of monitoring which was  
59 performed in seven residential neighborhoods throughout Wayne County, MI. The selection of  
60 neighborhoods and participants for study inclusion was based upon an a priori hypothesis that  
61 each neighborhood represented a potentially distinct air quality scenario being influenced by  
62 both regional as well as local pollution sources. Daily (24-hr integrated) measurement data were  
63 used to evaluate the spatial and temporal PM<sub>2.5</sub> compositional variability of the personal, indoor  
64 and outdoor spatial settings as they related to a central ambient monitoring site (Allen Park).  
65 Many of the PM<sub>2.5</sub> components were observed to have spatially different outdoor mass  
66 concentrations in matched neighborhood by neighborhood comparisons, with sulfate, OC, and  
67 NO<sub>3</sub> being noted exceptions. Coefficient of divergence (COD) comparisons involving outdoor  
68 measures for Ca, Si, Fe, Zn, Pb, and EC revealed significant spatial variability. While  
69 concentrations of most components were lower indoors as compared to outdoor measures, K and  
70 Si indoor concentrations often reflected aerosol enrichment (indoor/outdoor ratios  $\geq 1.2$ ). Even  
71 when personal exposures were adjusted for day to day changes in ambient concentrations, certain  
72 components (Ca, Fe, Mn, Zn, among others) revealed a high degree of location-specific spatial  
73 variability suggesting the influences of personal activities and/or local source influences on total  
74 personal PM<sub>2.5</sub> exposures. As a whole, findings indicate that reliance on a central ambient  
75 monitor as a surrogate for total personal and potentially even residential outdoor estimates of

76 PM<sub>2.5</sub> aerosol composition may provide an undesirable degree of exposure uncertainty for health-  
77 based risk estimates. The focus of this paper is on the spatial variability and uncertainty in using  
78 a central monitoring site to estimate exposures. Additional information concerning the DEARS  
79 can be found at <http://www.epa.gov/DEARS/>.

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## 81 **2.0 Introduction**

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83 Understanding the uncertainty of using a central (ambient) community monitoring site to  
84 estimate concentrations of air pollutant exposures for a given population or geographical area is  
85 of research interest to exposure scientists, environmental epidemiologists, and others involved in  
86 establishing health-based risk assessments (Dominici et al. 2006 [1], Brook et al 2011 [2]). Both  
87 spatial as well as temporal variability issues may be influencing factors on the reliability of using  
88 a central monitoring site as an adequate surrogate of a subject population's exposure. While the  
89 need to establish such potential measurement errors have been raised (Zeger et al. 2000[3];  
90 Navidi et al. 1994 [4]; Lipfert et al. 1997 [5]), few research studies have been specifically  
91 designed to examine this issue. The focus of this paper is on the spatial variability and  
92 uncertainty in using a central monitoring site to estimate exposures. The correlation of the central  
93 monitoring site to specific micro-environments (indoor, outdoor) and human exposures are the  
94 basis for these evaluations. The compositional components that are correlated are sulfates (SO<sub>4</sub>),  
95 nitrates (NO<sub>3</sub>), organic carbon (OC), elemental carbon (EC) and some of the crustal materials  
96 (Fe, Si, Ca, K, Mn, Pb, Zn).

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98 One such effort to obtain sufficient data to examine exposure measurement uncertainty in a given  
99 geographical area has recently been completed. The U.S. EPA conducted an intensive 3-year

100 human observational exposure study entitled, the Detroit Exposure and Aerosol Research Study  
101 (DEARS). The study was conducted in Wayne County, MI from 2004 to 2007. The DEARS was  
102 designed to investigate the sources of different pollutants impacting households across a large  
103 metropolitan area and to determine the spatial and temporal variability of a wide range of  
104 pollutant species at the personal (P), residential indoor (I), and residential outdoor (O), settings.  
105 The DEARS involved 24 hr-integrated (daily) monitoring associated with 142 participants and  
106 involving six selected neighborhoods. These six neighborhoods or enumeration monitoring areas  
107 (EMAs) were selected a priori as potentially being impacted by a wide range of both regional  
108 and local air quality sources. Summer and winter sampling schemes for each participant  
109 consisted of 5 days of monitoring each season (Tuesday – Saturday). In addition, daily pollutant  
110 measurements were taken at a centrally-located ambient monitoring site (A) at Allen Park, MI.  
111 Data from a total of three summer and three winter seasons were collected (Williams et al.  
112 2009[6]; EPA 2012 [7]). DEARS investigated the intra-urban variability in air pollution source  
113 impacts using receptor and statistical modeling of daily speciated PM<sub>2.5</sub> and VOC measurements  
114 collected at residential outdoor locations across Wayne County, MI (Duvall et al. 2012 [8];  
115 Bereznicki et al. 2012 [9]; George et al. 2010 [10]). Spatial relationships between coarse  
116 particulate matter in the DEARS were reported by Thornburg et al 2010 [11].

117  
118 Particulate matter (PM) represented one of the primary pollutants of interest in the DEARS with  
119 many reported PM pollutant sources present in the Detroit area. Wayne County, MI is  
120 consistently reported as one of the most polluted counties in the U.S. and the most polluted in  
121 Michigan as reported by the EPA’s Toxic Release Inventory (TRI). DEARS research observed  
122 negligible PM total mass (coarse) concentration spatially in residential outdoor measurements

123 across the Detroit urban air shed (Rodes et al. 2010 [12]). Spatial factors, such as distance from a  
124 highway, topography, land surface roughness, and the presence of other pollution sources affect  
125 the pollutant concentration and composition. Time-related factors, such as local meteorology  
126 (wind speed and direction, stability of the atmosphere boundary layer, precipitation, etc.), as well  
127 as traffic intensity may play a role in pollutant dispersion, and as a result in human exposure  
128 (Martuzeviciusa et al. 2004 [13]). George et al. 2010 [10] reported the spatiality influence of  
129 meteorology in neighborhood-based PM<sub>2.5</sub> mass concentrations associated with the DEARS.

130

131 Local PM<sub>2.5</sub> sources in Wayne County include industrial and residential combustion processes,  
132 motor vehicle emissions, residential and prescribed burning among a variety of others that  
133 contribute to the local air quality (Duvall et al. 2012 [8]; Bereznicki et al. 2012 [9]). PM<sub>2.5</sub> is  
134 formed from combustion processes and chemical reactions in the atmosphere and contains a  
135 wide variety of primary components. The major components of PM<sub>2.5</sub> are sulfates, nitrates,  
136 elemental/organic carbon (EC/OC), metals, and crustal elements. Some of these components  
137 have been reported to be associated with some negative health outcomes. Ostro et al. 2008 [14]  
138 found that cardiovascular mortality has been associated with PM<sub>2.5</sub> and several of its species  
139 including EC, OC, nitrates, sulfates, potassium, copper and iron. EC/OC has been associated  
140 with respiratory and cardiovascular health effects (Gauderman et al. 2004 [15]; Peters et al. 2000  
141 [16]). Sulfate (SO<sub>4</sub>) has advantages over other PM<sub>2.5</sub> components for retrospective epidemiology  
142 because extensive epidemiological literature and large databases for sulfates exist as compared to  
143 studies of the other components. The association of mortality with SO<sub>4</sub> is inconsistent. In a  
144 review of toxicologic studies, Schlesinger et al. (2003) [17] suggested that SO<sub>4</sub> is benign. In vivo  
145 studies PM<sub>2.5</sub>, Seagrave et al. (2006) [18] found that lung toxicity and inflammation correlated

146 with vehicular pollution but not secondary particles, including SO<sub>4</sub>. However, vehicular  
147 emissions are consistently associated with cardiac or other end points as reported by Grahame et  
148 al. (2007) [19]. Cavallari et al. 2008 [20] reports that the metal components of PM<sub>2.5</sub> may be  
149 toxic and responsible for lung inflammation and cardiac arrhythmias, and Valko et al. 2006 [21]  
150 reported that metal-induced toxicity and carcinogenicity are caused by oxidative stress.

151  
152 While specific PM mass components have been associated with health outcomes, little is known  
153 about the spatial and temporal variability of the mass concentrations of these components across  
154 a metropolitan area. Understanding such variability is critical in assessing the exposure  
155 measurement uncertainty or even the exposure misclassification errors in using a central  
156 community monitor to represent a given epidemiological study population (Zeger et al. 2000  
157 [3]). Significant sources of the trace metal and the crustal components of PM<sub>2.5</sub> in metropolitan  
158 settings may exist and could exhibit substantial spatial and temporal variability within such  
159 settings (Oglesby et al. 2000 [22]; Lau et al. 2009 [23]). It has been suggested that in such cases,  
160 centrally located community monitors might not be an adequate surrogate for residential  
161 concentrations and personal exposures to air pollutants (Kousa et al. 2002 [24]; Violante et al.  
162 2006 [25]). Examination of spatial and temporal variations in the concentration and composition  
163 of PM has the potential to provide important insights into particle sources and atmospheric  
164 processes that influence particle formation (Olofson et al. 1994 [26]; Motallebi et al. 2003 [27]).  
165 Investigations involving the seasonal and annual variability of the components of PM<sub>2.5</sub> would  
166 allow for the examination of the influence of the atmospheric contribution of a heavily  
167 industrialized urban center and the particulate matter composition (Ledoux et al. 2006 [28]).

168

169 To examine some of the issues discussed above relating to spatial and temporal variability of the  
170 major PM<sub>2.5</sub> mass components, we will report daily (24-hr) levels of personal, residential indoor  
171 and residential outdoor, as well as community-level concentrations of these components from the  
172 DEARS. A variety of statistical approaches are used in this assessment and extensive use of  
173 descriptive statistics, mixed models, and coefficient of divergence analyses provides the basis for  
174 summary findings.

175

176 Detroit is located in Wayne County, MI and EMAs selected for the sampling in DEARS are  
177 located within the county (Figure 1). Williams et al. 2009 [6] have described in great detail each  
178 EMA and their selection as part of the overall DEARS study design. In addition, preliminary  
179 investigations concerning potential industrial sources impacting the various EMAs have been  
180 reported (Duvall et al. 2012 [8]; Bereznicki et al. [9]). A selection of the EMAs was based on  
181 the proximity to point and line sources (local freeways or interstate highways) that were expected  
182 to impact these areas (Supplemental Table 1). The mobile sources are represented as a distance  
183 either less than or greater than 300 m from the roadway. The 300 m distance cut-off for roadway  
184 proximity is based on the hypothesis that concentrations of some mobile source-related  
185 pollutants (VOCs) decrease significantly at distances beyond 300 m from the source. Findings in  
186 the DEARS have supported this element of the study design (Barzyk et al. 2009 [29]).

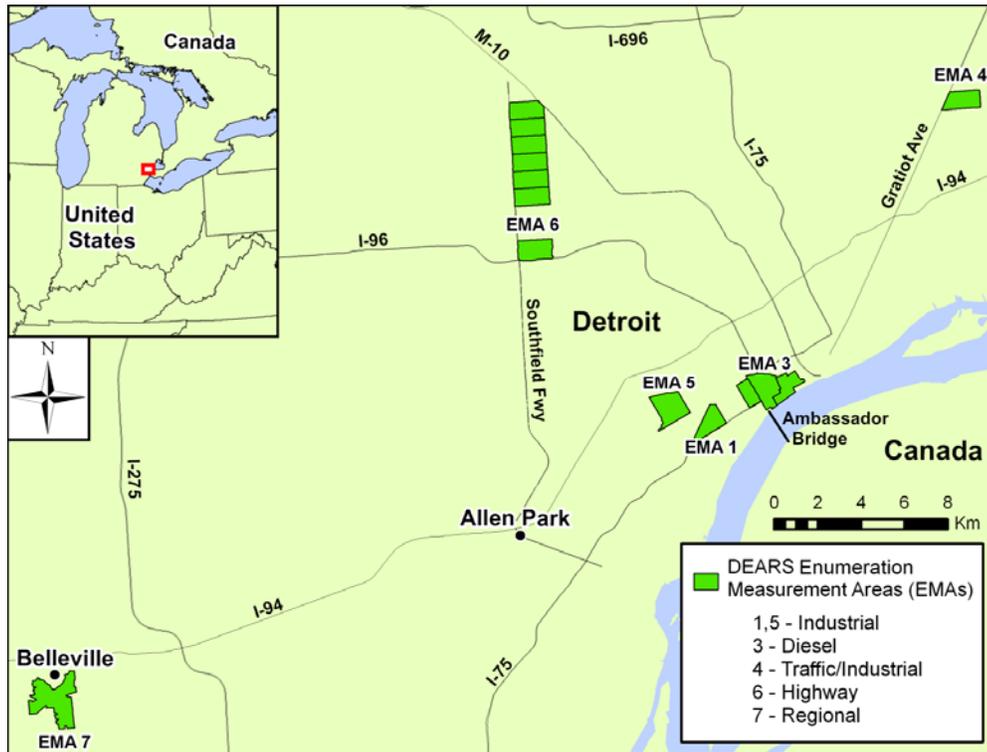
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188 EMA 1 represents the Zug Island area, a heavily industrialized island in the city of River Rouge  
189 near the southern city limits of Detroit. A major source of pollution in this area is from the steel  
190 manufacturing process. The Ambassador Bridge is believed to be a major PM source located in  
191 EMA 3. The bridge joins the US to Ontario, Canada and is North America's most active

192 international Border crossing. EMA 4 represents a mixture of both industrial as well as potential  
193 near-road impacts. Dearborn (EMA 5) is the center of the Detroit automotive industry. The  
194 EMAs includes six automotive factories on 600 acres (2.4 km<sup>2</sup>) of land, as well as steelmaking  
195 operations in the south end of Dearborn. A major source of air pollution in EMA 6 was  
196 hypothesized as the Southfield Freeway. The Michigan Department of Transportation (MDOT)  
197 surveys in 2010 showed that the highest traffic levels along the freeway were the  
198 159,400 vehicles daily between Schoolcraft Road and Grand River Avenue in Detroit; the lowest  
199 counts were the 20,400 vehicles per day between the I-94 and Van Born Road interchanges  
200 (MDOT 2010 [30]). Belleville (EMA 7) was considered a priori to be a background site  
201 impacted almost entirely by regional air quality. The central (ambient) monitoring site at Allen  
202 Park was collocated with one operated by the State of Michigan as part of their state  
203 comprehensive air monitoring network. This site has historically been used for compliance in  
204 demonstrating attainment with the National Ambient Air Quality Standards (NAAQS).  
205 Additional information concerning the DEARS can be found at <http://www.epa.gov/DEARS/>.  
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**Figure 1: Geographical map of DEARS Enumeration Monitoring Areas**



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### 210 3.0 Methods & Materials

211

#### 212 3.1 Study Design

213

214 Williams et al. 2009 [6] report on the design and field implementation for the DEARS. Personal  
215 samples were collected using active (2 lpm) PM<sub>2.5</sub> personal monitors (PEMs) placed on sampling  
216 vests worn by the participants. The residential samples, collected using the personal monitoring  
217 device, were collected outside of the homes, and the community (ambient) based monitoring  
218 took place at Allen Park, MI. EMAs were selected based on proximity to known or suspected  
219 point and line sources. Participants were monitored for 5 consecutive (24-h) days in each of two  
220 consecutive seasons (summer, winter) from 2004-2007. Selection criteria for participants were  
221 that they must be (1) non-smokers, (2) living in a non-smoking household, (3) ambulatory, (4)

222 expected to live in the same dwelling for the next 9 months, (5) living in a detached home, (6) 18  
223 years of age or older, and (7) able to comprehend either English or Spanish instructions. There  
224 were no health restrictions on enrollment other than being ambulatory. Likewise, there were no  
225 enrollment restrictions on occupation, socioeconomic status, sex, or ethnicity. Residences were  
226 selected using randomized sampling in a geographically focused pattern recruitment of  
227 participants (Phillips et al. 2010 [31]).

228

### 229 3.2 *Sample Collection and Analyses*

230 The measurements and analyses methods are generally described in Williams et al. 2009[6] and  
231 are referenced in the Supplemental Table 2.

232

### 233 3.6 *Data Analysis*

234

235 Descriptive statistics and distributions of PM<sub>2.5</sub> components were tabulated by season and  
236 EMAs. Descriptive analyses included the use of several measures of centrality (e.g., means and  
237 medians) and measures of dispersion (e.g., standard deviations and range of distributions) to  
238 characterize the distribution of the PM<sub>2.5</sub> components. Multivariate analysis included the use of  
239 mixed models to account for potential serial correlations between the repeated measurements.  
240 Multivariate analysis was performed using general linear models to examine the effect of  
241 seasonal variability on selected personal and outdoor elemental components. More specifically,  
242 we used the MIXED procedure in SAS (version 9.1) to account for potential serial correlations  
243 among repeated measures for each subject. The model was defined as:

244

245 
$$Y_{ij} = \beta_0 + \beta_1 X_{1ij} + \beta_2 X_{2ij} + \epsilon_{ij}$$

246

247  $i=1,\dots,k$  subjects,

248  $j=1,\dots,n_i$  measurement on the  $i^{\text{th}}$  subject

249

250  $Y_{ij}$  is the value of metal value associated with  $j^{\text{th}}$  measurement on the  $i^{\text{th}}$  subject

251

252  $\beta_0$ ,  $\beta_1$ , and  $\beta_2$  are parameters

253

254  $X_{1ij}$  is 0,1 variable representing two seasons

255

256  $X_{2ij}$  is the observed ambient value of the  $j^{\text{th}}$  measurement on the  $i^{\text{th}}$  subject

257

258  $\epsilon_{ij}$  is the random error for the  $j^{\text{th}}$  measurement o the  $i^{\text{th}}$  subject

259

260 The MIXED procedure requires a covariance structure to be specified in the model. We used the  
261 information criteria to produce the MIXED procedures as a tool in selecting a covariance for the  
262 model. We examined two covariance structures: compound symmetry (exchangeable) and  
263 autoregressive AR (1). After examining the two covariance structures, we chose the  
264 exchangeable covariance structure based on the Akaike Information Criteria (AIC) statistic. The  
265 AIC statistics associated with exchangeable covariance structure was smaller (Littell et al. [32])  
266 than the one associated with AR (1), and therefore the exchangeable covariance was used in the  
267 analyses. The exchangeable covariance structure indicated that correlations of the repeated  
268 measures were relatively constant. Within the mixed model, we also generated least square  
269 means for both seasons for selected components. In additional to examining potential  
270 differences between the two seasons, the least square means provided a magnitude of the

271 difference within-group means adjusted for other factors in the model. A statistical difference  
272 between variables being compared was reported when p-values were  $\leq 0.05$ .

273  
274 The Coefficient of Divergence (COD) between EMAs assessed spatiality of the  $PM_{2.5}$   
275 components (Pinto et al. 2004 [33]). In this study, we examined 6 EMAs and the central site,  
276 resulting in 21 pair-wise spatial comparisons. A COD of 0 indicates complete homogeneity and a  
277 value of 1 indicates maximum differences. COD values between 0 and 0.2 are representative of  
278 good agreement between matched pairs. On the contrary, values greater than 0.2 to 1 are  
279 indicative of pairs that do not agree well and are non-representative of one another. Enrichment  
280 factors presented in the paper are the mass concentration ratios calculated using the matched  
281 daily average means to estimate the relationships of indoor to outdoor or personal to indoor mass  
282 concentration relationships.

283

#### 284 **4.0 Results and Discussion**

285

286 The mean statistical summary of the primary  $PM_{2.5}$  mass components measured at the central  
287 (ambient) monitoring site for the summer and winter seasons in the DEARS observed that the  
288 total daily  $PM_{2.5}$  mass concentrations ranged from 2.8 to  $66.4 \mu\text{g}/\text{m}^3$  over the course of the full  
289 study. Using the accepted conversion factors for transforming elemental sulfur to sulfate ( $\text{SO}_4 = \text{S}$   
290  $\times 4.125$ ), the results indicate that the total  $PM_{2.5}$  mass is composed of  $\sim 36\%$   $\text{SO}_4$  in the summer.  
291 By contrast,  $\text{NO}_3$  was the major mass contributor observed during the winter seasons ( $\sim 29\%$ ).  
292  $\text{NO}_3$  exhibited the greatest seasonal difference in mass concentration than any other component  
293 with the mean winter concentration being more than 4 times that of the summer (Supplemental  
294 Table 3). Mean OC concentrations revealed little variability by season ( $\sim 1\%$ ). When original

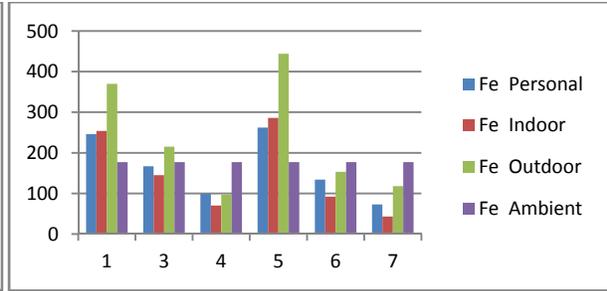
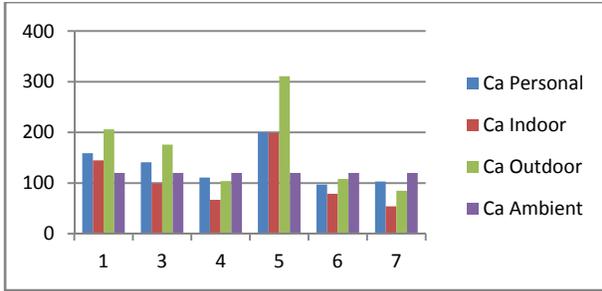
295 OC data was converted to its usual form for mass reporting (1.4 X OC), it contributed  
296 significantly to the total PM<sub>2.5</sub> mass (~23% in winter). Ca, Fe, K, Mn, Pb, Si, and Zn contributed  
297 significantly less mass to the total PM<sub>2.5</sub> composition regardless of season. Mean mass  
298 concentrations for these elements was typically  $\leq 200$  ng/m<sup>3</sup>. Even so, on some occasions they  
299 were observed to be significantly elevated on a daily basis (e.g., Fe maximum = 7130 ng/m<sup>3</sup>).  
300 Descriptive statistical data for personal, indoor, outdoor and the central site are found in the  
301 supplemental tables 3-9.

302  
303 Graphical representations (Charts 1 and 2) of the average concentrations of the data for the  
304 personal, indoor, outdoor and ambient concentrations of each component as they relate spatially  
305 for summer and winter show that the highest mass concentrations of PM<sub>2.5</sub> varies significantly  
306 within EMA 4 . The results for summer and winter show EMA 5 has the highest concentrations  
307 and greatest variance for the metals or crustal materials (Ca, Fe, K, Mn, Pb, Zn & Si) during  
308 summer and winter. EMA 5 (Dearborn) is a heavily industrialized area, and the most abundant  
309 metal was Fe contributions, averaging 921 ng/m<sup>3</sup>, were found there during the winter. Fe is the  
310 most abundant of the metals and is primarily associated with the soil and crustal elements of  
311 PM<sub>2.5</sub>. Duvall et al. 2011 [8], related the impact of a variety of steel manufacturing and mixed  
312 industries in the DEARS as a source of the observed Fe concentrations, especially those  
313 associated with EMA 1 and 5.

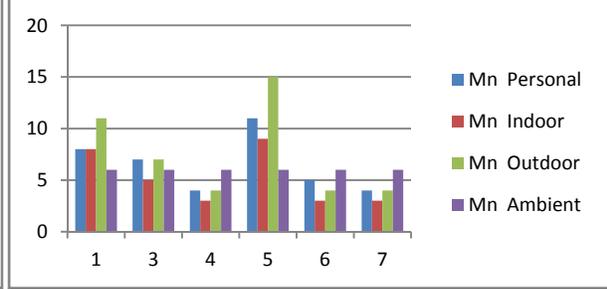
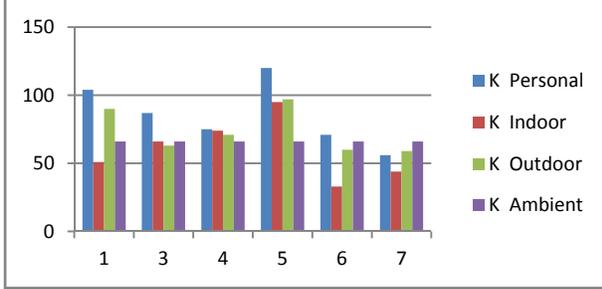
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315 **Chart 1. Component Mean Concentrations (ng/m<sup>3</sup>) in each Enumeration Monitoring Area (EMA)**  
316 **Personal, Indoor, Outdoor and Central Site (Summer)**  
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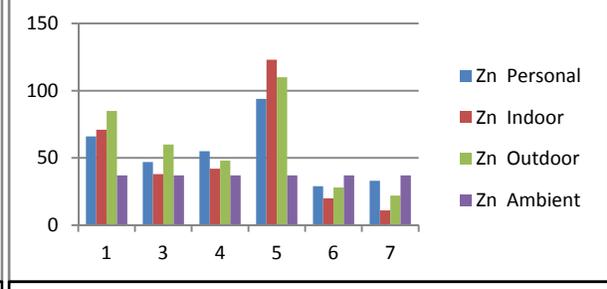
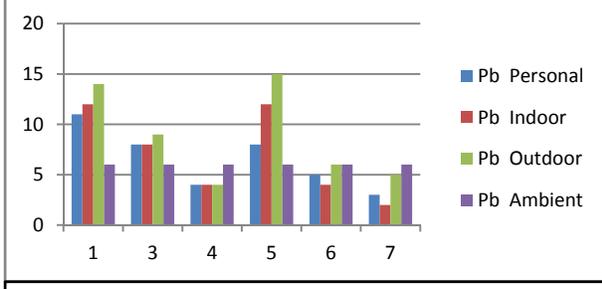
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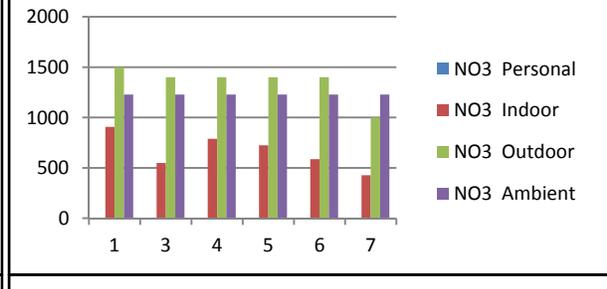
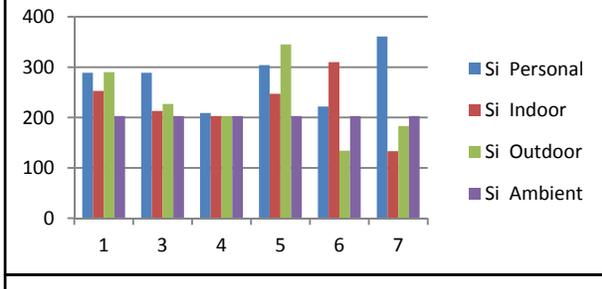
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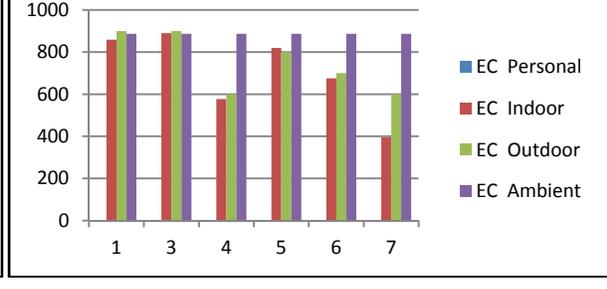
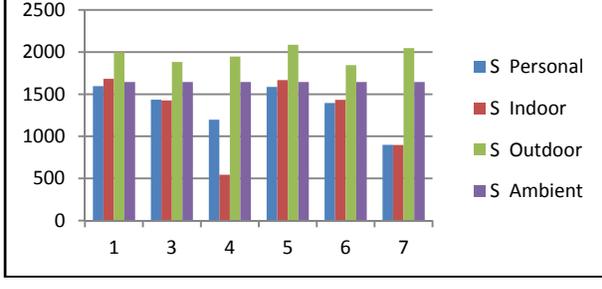
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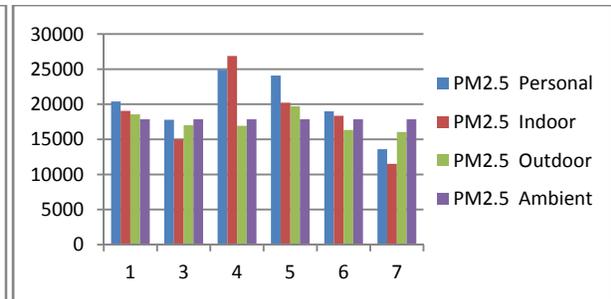
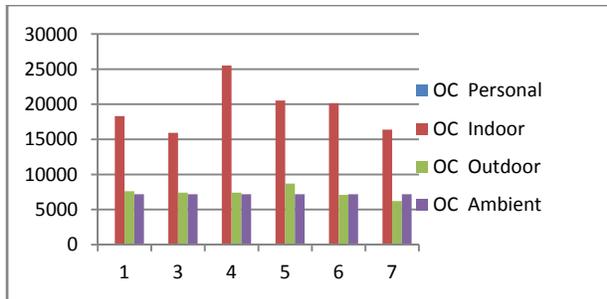


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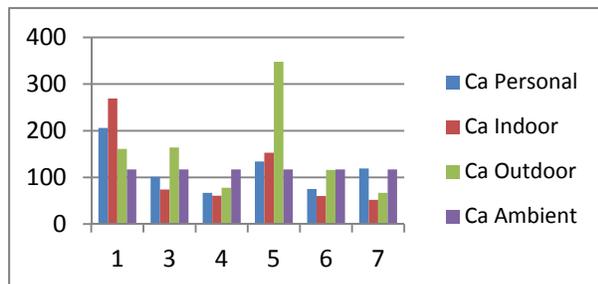
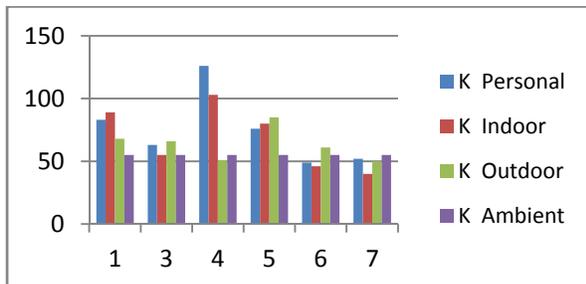
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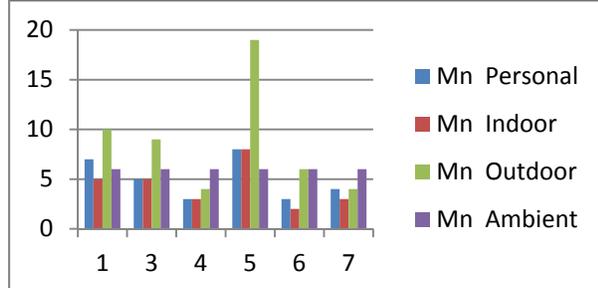
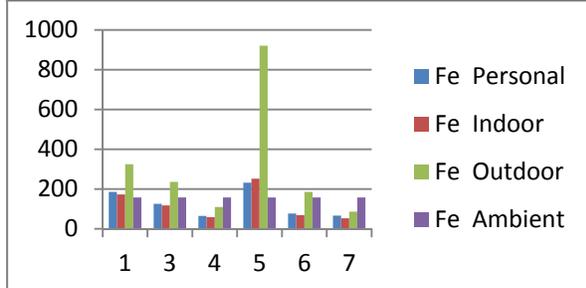


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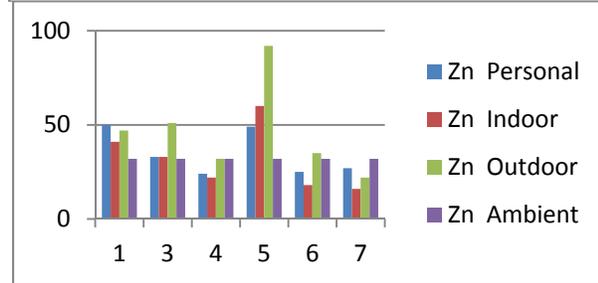
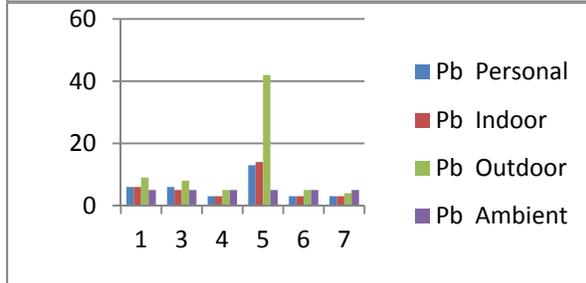
**Chart 2. Component Mean Concentrations (ng/m<sup>3</sup>) in each Enumeration Monitoring Area (EMA) Personal, Indoor, Outdoor and Central Site (Winter)**



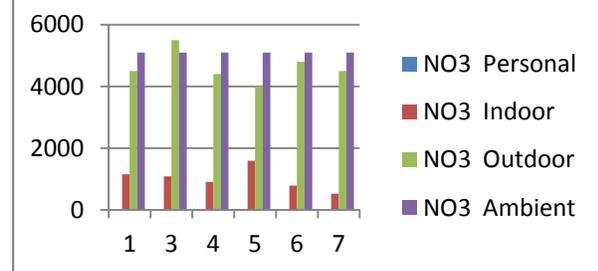
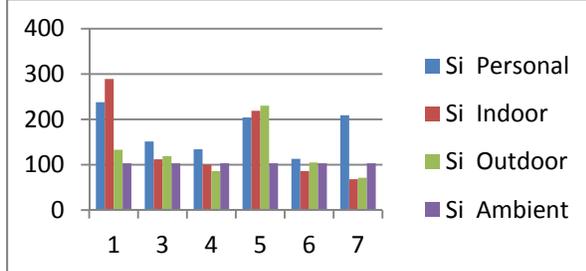
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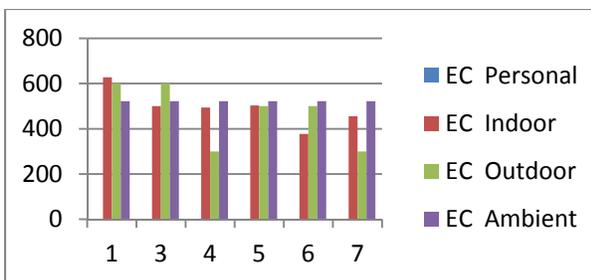
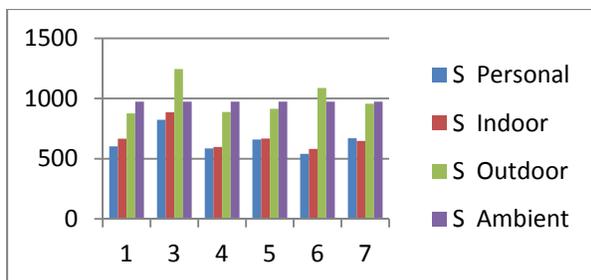


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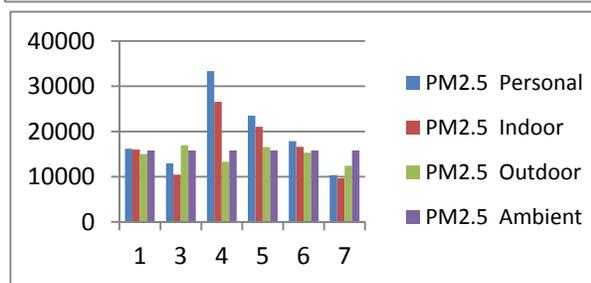
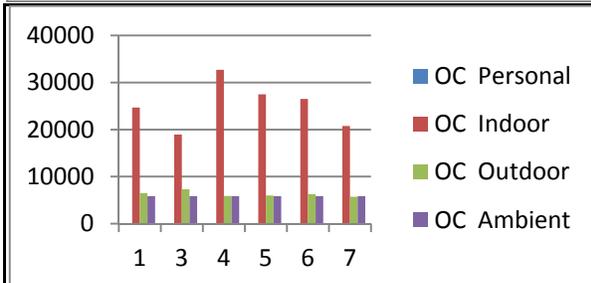


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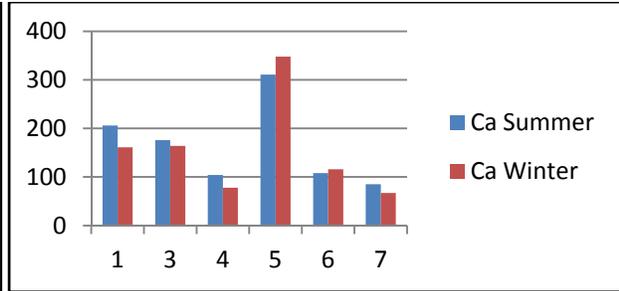
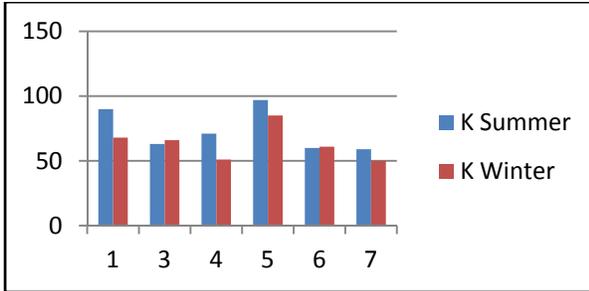


338 In general, total PM<sub>2.5</sub> mass across the EMAs was dominated by contributions from SO<sub>4</sub>, OC and  
 339 NO<sub>3</sub>. Winter data reveal that mean total PM<sub>2.5</sub> mass concentrations were highest in EMAs 3 and  
 340 5 (16.9 and 16.6 µg/m<sup>3</sup>, respectively) and EMAs 1 and 5 were highest in the summer  
 341 (supplemental tables 4 and 5). OC is the most abundant component for the outdoor residential  
 342 spatial setting with the highest concentrations observed in the summer. These findings are  
 343 consistent with reported findings that organic compounds of biogenic and anthropogenic origin  
 344 often represent a large fraction, up to 40%, of total PM mass (Chow et al. 1993 [34]; Chow et al.  
 345 1994 [35]). SO<sub>4</sub> is the second most abundant component in summer across the EMAs with NO<sub>3</sub>  
 346 being the second most abundant in winter. OC, SO<sub>4</sub> and NO<sub>3</sub> are considered secondary or  
 347 regional components of PM<sub>2.5</sub>. The graphical representations for the seasonal variations are  
 348 shown in Chart 3.

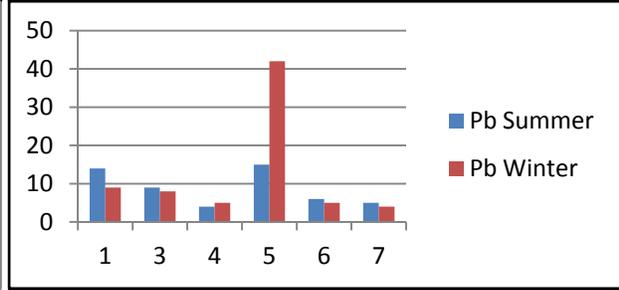
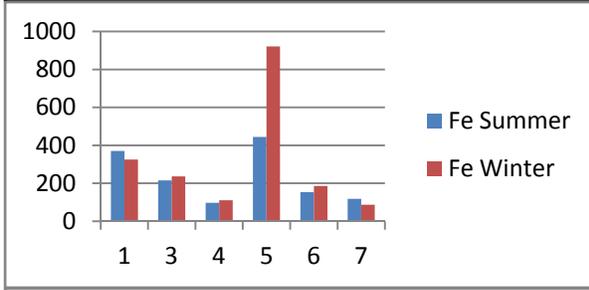
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Chart 3. Residential Outdoor Mean Concentrations (ng/m<sup>3</sup>) in each Enumeration Monitoring Areas (Summer vs Winter)

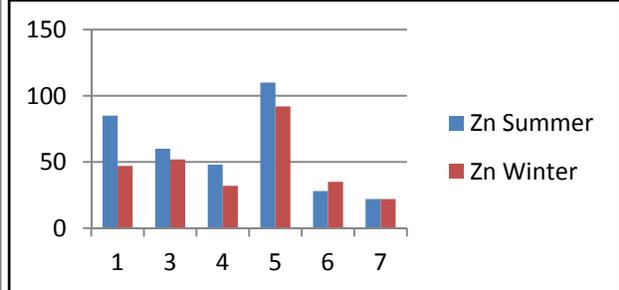
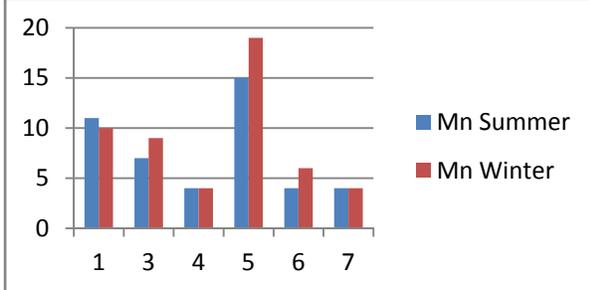
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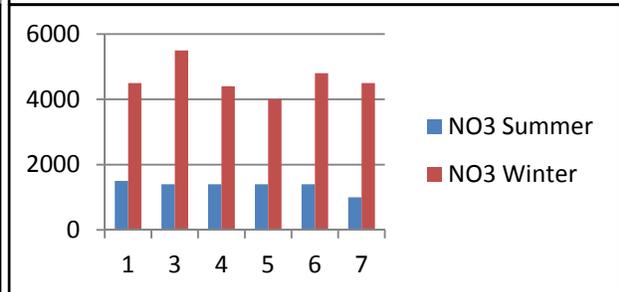
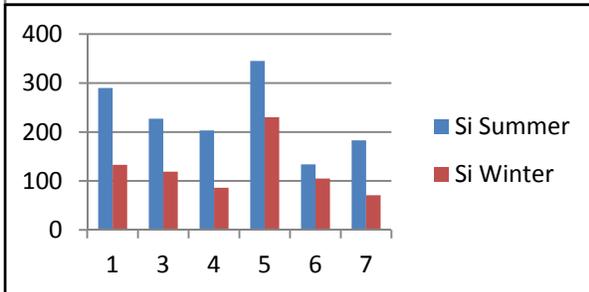
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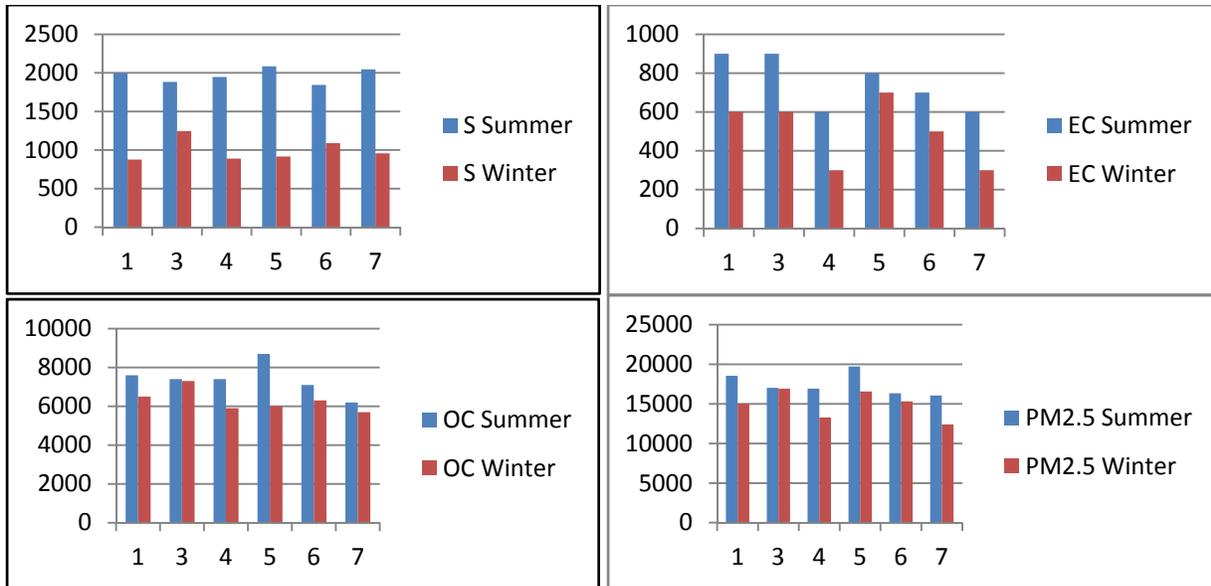
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Mixed model results indicate the impact of residential outdoor spatial PM<sub>2.5</sub> component

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variability across the DEARS EMAs by seasons (Table 1). Day to day variability of ambient-

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based concentrations was accounted for in the modeling approach. The presence of a p-value ≤

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0.05 for any of the components for a given season is indicative that some significant degree of

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spatiality exists. Such a value indicates that at least one of the EMAs had observed mean mass

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concentrations statistically different than the overall modeled mean. Rodes et al. 2010 [12] had

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identified some preliminary findings indicating that some minor PM<sub>2.5</sub> total mass heterogeneity

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existed across the DEARS on a daily basis (on the order of 1-2 μg/m<sup>3</sup>). The current findings

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validate that conclusion and provide for an EMA basis for such an observation. Using periodic

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sampling outdoor measures of S revealed low spatial variability. NO<sub>3</sub> and OC exhibited low

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spatial variability during the winter seasons. Again, as regional pollutants, such a finding of

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general homogeneity is not unexpected. What is surprising is the consistent pattern of some

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degree of heterogeneity that exists for the elemental components as a whole. This finding

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indicates that local sources of the various elemental components exist and are impacting the air

377 quality in one or more of the EMAs being compared. It further suggests that attempts to use a  
378 common ambient monitor to reflect neighborhood outdoor mass concentrations of select PM  
379 components of health interest (e.g., Fe, Zn, Mn) in epidemiological risk assessments may  
380 unknowingly introduce a high degree of exposure error. Considering that some of the DEARS  
381 EMAs were relatively close to one another ( $\leq 5$  km distance) and that some degree of overall  
382 spatiality was still observed for many of the elements, proximity of an ambient monitor to a  
383 target population (nearby location), may not be a sufficient decision parameter alone in  
384 conducting research of that nature.

<b>Table 1: MIXED Model Results – Examining the effect of spatial variability by season on outdoor element measures (ng/m<sup>3</sup>) using average mean concentrations.</b>								
Component	Season	Enumeration Monitoring Areas (EMAs)						p-Value
		1	3	4	5	6	7	
Calcium	Summer	208	171	75	300	111	98	< 0.01
	Winter	163	165	77	338	115	73	< 0.01
Iron	Summer	376	207	109	444	153	126	< 0.01
	Winter	331	230	115	902	183	98	< 0.01
Potassium	Summer	91	70	62	97	60	59	< 0.01
	Winter	74	61	53	86	58	52	< 0.01
Manganese	Summer	11	7	5	15	4	5	< 0.01
	Winter	10	9	4	19	6	4	< 0.01
Lead	Summer	12	8	6	15	6	5	< 0.01
	Winter	9	7	5	42	5	4	< 0.01
Zinc	Summer	83	61	29	107	28	22	< 0.01
	Winter	49	48	31	94	34	25	< 0.01
Silicon	Summer	288	247	201	344	195	205	0.003
	Winter	132	119	86	229	105	67	< 0.01
Nitrates	Summer	1487	1335	1294	1425	1342	1057	< 0.01
	Winter	4982	5085	4841	4834	5041	4178	0.11
*Sulfur	Summer	1999	1850	1945	2086	1846	1875	0.95

	Winter	889	1239	883	926	1077	1146	0.76
EC	Summer	915	945	586	835	719	542	< 0.01
	Winter	587	598	349	591	459	326	< 0.01
<sup>b</sup> OC	Summer	7657	7407	7304	8488	7153	6071	< 0.01
	Winter	6680	6537	5836	6381	6460	5605	0.15
PM <sub>2.5</sub>	Summer	18611	17146	15248	19610	16242	15003	< 0.01
	Winter	16200	16305	13403	18727	15017	12075	< 0.01

<sup>a</sup> Conversions used in the manuscript to quantify SO<sub>4</sub>: SO<sub>4</sub> = S x 4.125

<sup>b</sup> Conversions used in the Table OC = OC x 1.4

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389 We further identify the inter-EMA comparability of the PM<sub>2.5</sub> mass components (Table 2).  
390 Coefficient of divergence (COD) statistic is provided for the combined summer and winter  
391 measures. These measures provide a clear perspective on how residential outdoor measures in  
392 any one EMA compared to date-matched measures in all other EMAs and the central site. While  
393 there is not consensus of an exact COD value that constitutes statistical significance, literature  
394 indicates values > 0.2 are indicative of the pairings that are somewhat not representative of each  
395 other (Thornburg et al. 2010 [11]). Using such a threshold indicator of heterogeneity, the  
396 regional nature of NO<sub>3</sub>, S (SO<sub>4</sub>), and OC is clearly established where most of the pairing are  
397 approximately 0.2 or less. These components EC, typically associated with automotive and other  
398 similar fossil fuel combustion sources, exhibited greater spatial variability as many of the  
399 pairings exhibited CODs > 0.30. This observed wide-spread heterogeneity strengthens the  
400 earlier statistical finding associated with EC in Table 1. K on the other hand, exhibits  
401 significantly less spatial variability with a majority of the pairings having COD values ≤ 0.25. K  
402 can be considered to have low spatial and temporal variability when the individual EMAs are  
403 correlated using continuous central site monitoring over all seasons. K has been considered a  
404 possible regional source but its origin has not been accounted for in the Detroit area (Duvall et al.

405 2012 [8]). Residential outdoor concentration pairings for Ca, Si, Mn, Zn, and Pb were routinely  
 406 different across most of the EMA pairings (low spatial and temporal variability). EMA 7, the  
 407 regional background site, often exhibited a concentration difference relative to the more  
 408 metropolitan based EMAs (1, 3, 4, 5, and 6) with respect to elemental components. This is not  
 409 surprising considering the lack of industrial and other identifiable sources in that location.

**Table 2: Coefficient of Divergence (COD) between EMAs (winter & summer)**

Component	1 vs 3	1 vs 4	1 vs 5	1 vs 6	1 vs 7	1 vs 9	3 vs 4	3 vs 5	3 vs 6	3 vs 7	3 vs 9	4 vs 5	4 vs 6	4 vs 7	4 vs 9	5 vs 6	5 vs 7	5 vs 9	6 vs 7	6 vs 9	7 vs 9
NO <sub>3</sub>	0.14	0.14	0.21	0.17	0.22	0.21	0.15	0.16	0.14	0.22	0.16	0.21	0.14	0.19	0.22	0.19	0.22	0.14	0.22	0.22	0.21
S	0.06	0.12	0.07	0.13	0.12	0.10	0.09	0.05	0.12	0.12	0.08	0.11	0.10	0.13	0.13	0.12	0.11	0.12	0.08	0.11	0.13
K	0.20	0.27	0.21	0.25	0.26	0.24	0.20	0.25	0.21	0.26	0.21	0.33	0.20	0.22	0.21	0.31	0.32	0.21	0.31	0.18	0.20
Ca	0.23	0.46	0.28	0.37	0.48	0.33	0.40	0.36	0.33	0.48	0.21	0.60	0.27	0.26	0.31	0.51	0.62	0.33	0.47	0.29	0.26
Si	0.40	0.44	0.29	0.70	0.52	0.48	0.73	0.38	0.36	0.52	0.44	0.48	0.44	0.55	0.46	0.47	0.53	0.36	0.46	0.86	0.41
Mn	0.35	0.50	0.34	0.44	0.53	0.43	0.40	0.43	0.37	0.54	0.38	0.57	0.41	0.51	0.44	0.57	0.64	0.37	0.54	0.40	0.34
Fe	0.35	0.51	0.39	0.39	0.56	0.42	0.33	0.43	0.27	0.56	0.31	0.61	0.28	0.33	0.36	0.51	0.65	0.27	0.51	0.37	0.29
Zn	0.32	0.48	0.38	0.50	0.52	0.44	0.37	0.42	0.40	0.52	0.37	0.54	0.39	0.43	0.41	0.57	0.62	0.40	0.51	0.35	0.37
Pb	0.40	0.52	0.42	0.46	0.86	0.52	0.48	0.44	0.99	0.86	0.67	0.54	0.37	0.78	0.55	0.54	0.63	0.99	0.54	0.75	0.93
EC	0.21	0.37	0.29	0.30	0.42	0.26	0.32	0.30	0.28	0.42	0.22	0.33	0.24	0.33	0.31	0.28	0.42	0.28	0.28	0.42	0.27
OC	0.15	0.16	0.14	0.15	0.21	0.16	0.14	0.14	0.15	0.21	0.15	0.15	0.15	0.51	0.16	0.15	0.21	0.15	0.15	0.21	0.15
PM <sub>2.5</sub>	0.11	0.16	0.13	0.14	0.20	0.12	0.14	0.15	0.13	0.20	0.11	0.21	0.13	0.16	0.14	0.18	0.22	0.13	0.16	0.16	0.12

410 \*(values greater than 0.2 are indicative of pairs that are non-representative of one another. The central site is represented as Enumeration Monitoring Areas  
 411 (EMA) 9.)  
 412 \* Conversions used in the manuscript to quantify SO<sub>4</sub>: SO<sub>4</sub> = S x 4.125, Conversions used in the Table OC = OC x 1.4  
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414  
 415 We have previously reported that the time activity diaries for the DEARS participants show that  
 416 approximately 80% of their time is spent indoors at the residence (Rodes et al. 2010 [12]).  
 417 Personal exposures to particles are frequently dominated by exposure to non-ambient particles  
 418 and originate from indoor sources. Therefore, understanding how well residential indoor mass  
 419 concentrations of these PM components relate to ambient measures is critical in reducing

420 exposure uncertainty. Indoor PM<sub>2.5</sub> component concentrations revealed a high degree of  
 421 variability when compared to those from the ambient monitoring site (Table 3). A p value ≤ 0.05  
 422 is once again indicative of some degree of mass concentration heterogeneity associated with the  
 423 mean mixed model value for the PM component across all EMAs when adjusted for the day to  
 424 day variability of ambient-based mass concentrations. Seasonal residential indoor PM<sub>2.5</sub> mass  
 425 concentrations were observed to range from 8.8 to 31.5 μg/m<sup>3</sup>. Indoor PM<sub>2.5</sub> total mass  
 426 concentration associated with participants from EMA 7 represented the lowest means observed  
 427 regardless of season. Some degree of Fe, Mn, Pb, Zn, NO<sub>3</sub>, OC, and S (SO<sub>4</sub>) indoor mass  
 428 spatiality occurred over both the summer and winter seasons. While there are numerous indoor  
 429 sources of OC (cooking aerosols being one example) and thus a ready explanation for the  
 430 observed spatial effect, the observed spatiality for indoor S needs to be explained. The indoor  
 431 OC and S have high spatial and temporal variability using the periodic central site monitoring for  
 432 these evaluations. While it is a regional pollutant, we have identified environmental tobacco  
 433 smoke in the participant's homes as being an influencing factor on overall indoor S  
 434 concentrations in the DEARS (Williams et al., 2012 [ 36]), and thus the effect observed here. K  
 435 was the only component not observed to exhibit some degree of indoor residential statistical  
 436 significant or low spatial and temporal (p= 0.6) variability, although near significance was  
 437 observed for the winter season. Ca was significantly different during the summer.

**Table 3: MIXED Model Results – Examining the effect of spatial variability indoor element measures (ng/m<sub>3</sub>).**

Component	Season	Enumeration Monitoring Areas (EMA)						p-Value
		1	3	4	5	6	7	
Calcium	Summer	145	100	67	196	80	59	< 0.01
	Winter	262	74	59	146	60	54	0.17
	Summer	254	143	71	284	91	46	< 0.01

<b>Iron</b>	<b>Winter</b>	177	110	63	257	69	47	< 0.01
	<b>Summer</b>	86	68	75	99	66	43	0.13
<b>Potassium</b>	<b>Winter</b>	97	50	136	80	45	37	0.06
	<b>Summer</b>	7	5	3	9	3	3	< 0.01
<b>Manganese</b>	<b>Winter</b>	6	5	3	8	2	2	< 0.01
	<b>Summer</b>	10	8	4	12	4	2	< 0.01
<b>Lead</b>	<b>Winter</b>	6	5	3	14	3	2	< 0.01
	<b>Summer</b>	68	41	44	117	21	17	< 0.01
<b>Zinc</b>	<b>Winter</b>	43	31	23	61	18	14	< 0.01
	<b>Summer</b>	262	190	194	263	322	101	0.59
<b>Silicon</b>	<b>Winter</b>	288	112	99	218	86	68	< 0.01
	<b>Summer</b>	905	565	755	735	575	380	< 0.01
<b>Nitrates</b>	<b>Winter</b>	1214	888	1110	2186	755	346	0.02
	<b>Summer</b>	1680	1458	1308	1584	1440	1008	< 0.01
<b><sup>a</sup>Sulfur</b>	<b>Winter</b>	802	755	664	762	572	493	< 0.01
	<b>Summer</b>	861	904	576	807	674	411	< 0.01
<b>EC</b>	<b>Winter</b>	723	453	602	368	351	366	0.33
	<b>Summer</b>	18525	15000	25239	20280	20130	17117	< 0.01
<b><sup>b</sup>OC</b>	<b>Winter</b>	24244	18749	34921	27147	24457	20245	0.01
	<b>Summer</b>	19636	15067	25234	20245	18474	12054	0.02
<b>PM<sub>2.5</sub></b>	<b>Winter</b>	17786	9756	31573	26411	15538	8797	0.02

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<sup>a</sup> Conversions used in the manuscript to quantify SO<sub>4</sub>: SO<sub>4</sub> = S x 4.125

<sup>b</sup> Conversions used in the Table OC = OC x 1.4

442 We compared matched residential indoor versus residential outdoor PM<sub>2.5</sub> composition ratios  
443 (Supplemental table10). Such ratios are often considered as enrichment factors when ratios  
444 exceed unity (> 1.0). Total PM<sub>2.5</sub> indoor/outdoor ratios 1:1 indicate the significant contribution to  
445 the infiltration of outdoor air had on total mass concentrations as a whole. Even so, it must be  
446 realized that indoor sources of PM<sub>2.5</sub> also contributed to the totals. We have previously reported  
447 that the mean residential PM<sub>2.5</sub> infiltration factor in the DEARS was ~0.7 (Williams et al., 2009

448 [6]). Therefore, it is suggested that on average, residential indoor sources contributed ~ 30% of  
449 the total PM<sub>2.5</sub> mass observed and the resulting 1:1 ratios. While a clear majority of the  
450 comparisons had lower indoor concentrations of any respective component, some enrichment  
451 was observed. This was most notable for Ca (winter), K (summer and winter), Zn (summer), and  
452 Si (summer and winter). One might speculate on a variety of either indoor or indoor  
453 infiltrated/deposited sources responsible for each of those named immediately above. Descriptive  
454 statistics detail the residential indoor PM<sub>2.5</sub> components variability across the summer and winter  
455 seasons. The large degree of variability (often exceeding 100%) as measured by the RSD across  
456 the various components and by EMA suggests the difficulty that might exist in trying to  
457 associate ambient-based measures of these pollutants as surrogates for indoor concentrations in  
458 most instances. Future work will attempt to associate residential indoor and outdoor  
459 concentrations of these elements along with survey information obtained in the DEARS to  
460 investigate potential influencing human and environmental exposure factors.

461  
462 Data reported in Table 4 examines the effect of spatial variability on personal measures after  
463 adjustment for day to day variability of ambient-based measures. As can be seen in  
464 Supplemental Table 11, matched personal and residential indoor component mass concentration  
465 ratios were often within 20% of unity. This is not surprising considering the time activity pattern  
466 of the DEARS participants indicated a significant (~ 75%) amount of time spent home indoors  
467 each day (Rodes et al. 2010, [12]). Therefore, the residential indoor environment would have the  
468 largest time opportunity to influence the total daily personal exposure profile. Mn and Zn  
469 exhibited the greatest divergence from unity, and are suggestive of non-residential indoor source  
470 impacts on some of the participants. The p-value statistics ( $p \leq 0.05$ ) reported in supplemental

471 Table 12 indicates that both spatial and temporal effects are evident relative to ambient-adjusted  
472 personal exposures. In other words, the day to day variability observed in personal exposure  
473 PM<sub>2.5</sub> mass component heterogeneity across the EMAs cannot be accounted for by changes in  
474 the ambient conditions alone. The least degree of heterogeneity or spatial variability was  
475 observed for Ca (winter), K (winter), Zn (summer), and Si (summer). The observed  
476 heterogeneity for S observed here would appear to be due to the much lower mass concentrations  
477 observed in EMA 7 with respect to the other EMAs. One possible explanation for this  
478 observance would be that EMA 7 is upwind of the majority of industrial emissions in the  
479 DEARS study area and therefore less impacted by secondary organic aerosol products (e.g., SO<sub>4</sub>)  
480 impacting total personal exposures.

<b>Table 4: MIXED Model Results – Examining the effect of spatial variability using average mean concentrations (ng/m<sub>3</sub>) of personal measures adjusting for ambient air by season.</b>								
Component	Season	Enumeration Monitoring Areas (EMA)						p-Value
		1	3	4	5	6	7	
Calcium	Summer	159	141	111	200	97	103	< 0.01
	Winter	199	103	66	128	74	119	0.178
Iron	Summer	246	167	99	262	134	73	< 0.01
	Winter	192	120	68	233	77	61	< 0.01
Potassium	Summer	104	87	75	120	71	56	0.040
	Winter	88	60	153	76	48	50	0.123
Manganese	Summer	8	7	4	8	5	3	< 0.01
	Winter	7	5	3	8	3	3	< 0.01
Lead	Summer	11	8	4	11	5	4	< 0.01
	Winter	6	6	3	13	4	3	< 0.01
Zinc	Summer	66	47	55	94	29	33	0.12
	Winter	51	31	26	51	25	24	0.02
Silicon	Summer	289	289	209	304	222	361	0.08
	Winter	240	151	132	205	111	208	0.03
Sulfur	Summer	1596	1435	1197	1586	1396	899	< 0.01
	Winter	742	707	638	743	535	523	0.02

PM <sub>2.5</sub>	Summer	20,400	17,800	24,900	24,100	19,000	13,600	0.031
	Winter	16,200	12,400	34,300	26,900	16,000	9,700	0.014

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<sup>a</sup> Conversions used in the manuscript to quantify SO<sub>4</sub>: SO<sub>4</sub> = S x 4.125

484 Information reported in the supplemental tables 8 and 9 provide spatial and temporal descriptive  
485 statistics of personal PM<sub>2.5</sub> mass component observed in the DEARS. These tables give more  
486 insight into the variability of the personal measures. The large values associated with the RSD  
487 are an indication of the high spatial and temporal variability of personal exposures to the various  
488 PM components across the various EMAs. Taken into context with data presented it is evident  
489 that local and certainly some indoor-related sources often play a large role in total personal  
490 exposures to these PM mass components. It has been reported that indoor particulates are  
491 generated or re-suspended from everyday activities such as cooking, dusting, vacuuming, etc.  
492 (Wallace et al. 1996 [37]). “The source strengths were found to be a function of the number of  
493 persons performing the activity, the vigor of the activity, the type of activity, and the type of  
494 flooring (Ferro et al. 2004 [38]).” The impact of indoor activities on total personal exposures  
495 mentioned above vary between households and individuals which explains the variability in  
496 exposure. In addition to indoor residential activities, microenvironments affect personal  
497 exposures. These microenvironments include workplaces, outdoor surroundings, personal cloud,  
498 etc. (Wallace et al. 1996 [37]). Landis et al. [39] have reported on possible non-ambient related  
499 personal activities that appear to influence total personal PM<sub>2.5</sub> component exposures.

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501 **5.0 Conclusion**

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503 The DEARS represented an extensive matched personal, residential indoor, residential outdoor,  
504 and ambient-based spatial and temporal study design and provided hundreds of PM<sub>2.5</sub> mass  
505 component comparison opportunities. It is evident in the data provided that attempting to use  
506 ambient measures as effective surrogates of exposures to specific PM<sub>2.5</sub> mass components might  
507 be problematic and could lead to substantial exposure measurement uncertainty or potentially  
508 even health outcomes misclassification in health-based risk assessments. Even adjusting for day  
509 to day changes in the ambient environment often failed to negate the observed differences at the  
510 residential outdoor, residential indoor, and especially at the personal level. Local (unknown)  
511 sources are impacting many of the EMAs investigated in the DEARS. The reports by Bereznicki  
512 et al. 2012 [9] and Duvall et al. 2012 [8] provide some insight as to these local source impacts.  
513 Examination of the extensive time activity and residential survey information obtained in the  
514 DEARS will now be used in future efforts to elucidate the specific activities that impacted the  
515 study population's exposure to non-ambient PM<sub>2.5</sub> mass components. Additional information  
516 concerning the DEARS can be found at <http://www.epa.gov/DEARS/>. EPA is working toward a  
517 web-based public release of the DEARS data in the future that holds the potential for  
518 collaboration on additional data analyses.

519

## 520 **6.0 Acknowledgements**

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<b>Supplemental Table 1: DEARS residential neighborhoods and possible source impacts</b>			
Enumeration Monitoring Areas no.	General location <sup>a</sup>	Potential source impact <sup>b</sup>	Neighborhood proximity to freeway <sup>c</sup>
<b>1</b>	Zug Island	Heavy industrial	≥ 300m
<b>3</b>	Ambassador Bridge	Diesel truck traffic	≤ 300m
<b>4</b>	US 3 and 7 Mile Road	Industry and automotive	≤ 300m and ≥ 300m
<b>5</b>	Dearborn	Industrial	≥ 300m
<b>6</b>	Southfield Freeway	Mobile source (freeway)	≤ 300m
<b>7</b>	Belleville	Regional	≥ 300m
<b>Allen Park</b>	Southern Wayne County	Industrial and freeway	≤ 300m

<sup>a</sup> Represents the general area of the neighborhood sampled

<sup>b</sup>Source impacts defined here are those believed from a *priori* determinations of the National Emissions Inventory (NEI) and other information gathered before the study was initiated.

<sup>c</sup> Neighborhoods within 300m of busy roadways were considered as potentially impacted by near-road mobile source emissions.

**Supplemental Table 2: DEARS sample collection and analysis methods.**

<b>Metric</b>	<b>Type</b>	<b>Field collection</b>	<b>Analysis</b>	<b>Analysis reference</b>
PM2.5 mass	P, I, O, A	2 l/m PEM filter based	Gravimetric	Lawless and Rodes (1999)
PM2.5 elements	P, I, O, A	2–4 l/m PEM Teflon filter	ED-XRF	Dzubay et al. (1988)
PM2.5 EC–OC	I, O, A	2 l/m PEM quartz filter	Thermal–optical reflectance	Birch and Cary (1996)
PM2.5 nitrate	I, O, A	0.8 l/m Coat denuder/quartz filter	IC	Demokritou et al.(2001)
NO <sub>2</sub> , O <sub>3</sub> , SO <sub>2</sub>	I (NO <sub>2</sub> )P, A	Ogawa passive diffusion	IC	Varns et al. (2001)

P = personal, I = indoor, O = outdoor, A = ambient, PEM= personal exposure monitor, ED-XRF = energy dispersive x-ray fluorescence, IC = ion chromatography

**Supplemental table 3: Mean Descriptive Statistics for the Central Monitoring Site**  
Summer/Winter

<b>Component</b>	<b>N Observations</b>	<b>Mean (ng/m<sup>3</sup>)</b>	<b>Std Dev</b>	<b>RSD %</b>	<b>Minimum (ng/m<sup>3</sup>)</b>	<b>95<sup>th</sup> Pctl(ng/m<sup>3</sup>)</b>	<b>Max(ng/m<sup>3</sup>)</b>
<b>Calcium</b>	100 / 70	120 / 117	54 / 74	45/63	39 / 24	400 /348	957 / 1916
<b>Iron</b>	100 / 70	177 / 158	74 / 98	42/62	31 / 46	637 / 748	2526 / 7130
<b>Potassium</b>	100 / 70	66 / 55	31 / 32	46/58	18 / 12	155 / 145	390 / 385
<b>Manganese</b>	100 / 70	6 / 6	3 / 4	54/80	1 / 1	20 / 23	56 / 77
<b>Lead</b>	100 / 70	6 / 5	4 / 4	68/87	1 / 0	21 / 21	158 / 665
<b>Zinc</b>	103 / 70	37 / 32	37 / 34	100/107	3 / 4	171 / 132	1262 / 333
<b>Silicon</b>	100/91	203/103	312/79	154/77	0/0	362/236	2732/519
<b>Nitrate</b>	102/95	1228/5097	847/4878	69/96	0/0	2437/13973	5513/27822
<b>Sulfur</b>	100/91	2080/974	1644/799	79/82	241/198	5672/2749	6792/4487
<b>EC</b>	104/95	887/522	299/273	34/52	380/0	1420/942	1723/1667
<b>OC</b>	104/95	7170/5842	2637/2277	37/39	1030/2351	12131/9046	15231/14847
<b>PM<sub>2.5</sub></b>	<b>98/91</b>	<b>17850/15808</b>	<b>9806/11397</b>	<b>55/72</b>	<b>3337/2787</b>	<b>40305/37163</b>	<b>43741/66371</b>

\*Conversions used in the manuscript to quantify SO<sub>4</sub>: SO<sub>4</sub> = S x 4.125,

\* Conversions used in the Table OC = OC x 1.4

**Supplemental Table 4: Residential Outdoor Concentrations in the Enumeration Monitoring Areas (Summer)**

Enumeration Monitoring Areas	Component	N Observations	Mean (ng/m <sup>3</sup> )	Std Dev (ng/m <sup>3</sup> )	RSD %	Minimum (ng/m <sup>3</sup> )	95 <sup>th</sup> Pctl (ng/m <sup>3</sup> )	Max (ng/m <sup>3</sup> )
1	Calcium	122	206	109	53	47	406	641
	Iron	122	370	248	67	81	889	1217
	Potassium	122	90	58	65	25	207	390
	Manganese	122	11	6	58	2	25	31
	Lead	122	14	18	136	1	29	158
	Zinc	122	85	100	118	10	266	712
	Si	122	290	338	117	<MQL	575	3599
	NO <sub>x</sub>	121	1500	1200	80	200	3600	5300
	S	122	1997	1640	82	184	5501	6779
	EC	126	900	300	33	300	1400	1700
	OC	126	7600	2600	34	2000	13000	16000
PM <sub>2.5</sub>	121	<b>18554</b>	<b>9800</b>	<b>53</b>	<b>4332</b>	<b>37428</b>	<b>55581</b>	
3	Calcium	167	176	109	62	24	375	957
	Iron	167	215	158	73	47	447	1546
	Potassium	167	69	33	48	22	138	231
	Manganese	167	7	4	61	<MQL	13	32
	Lead	167	9	6	73	0.0	20	49
	Zinc	167	60	101	167	6	204	825
	Si	167	227	151	67	18.6	493	1168
	NO <sub>x</sub>	152	1400	900	64	40	1900	3100
	S	167	1883	1471	78	191	5183	7449
	EC	168	900	400	44	300	1600	2100
	OC	168	7400	2400	32	1800	11600	15000
PM <sub>2.5</sub>	167	<b>17026</b>	<b>8677</b>	<b>51</b>	<b>4686</b>	<b>34004</b>	<b>43445</b>	
4	Calcium	107	104	201	192	12	192	1757
	Iron	107	97	101	105	13	219	797
	Potassium	67	71	76	106	<MQL	166	556
	Manganese	108	4	4	119	<MQL	10	31
	Lead	67	4	5	124	<MQL	11	23
	Zinc	109	48	155	323	<MQL	100	1173
	Si	127	203	270	133	<MQL	2100	499
	NO <sub>x</sub>	124	1400	1000	71	200	3400	5100
	S	127	1948	1535	79	65.5	5249	6875
	EC	129	600	200	33	150	900	1700
	OC	129	7400	2500	34	1500	12000	13600
PM <sub>2.5</sub>	124	<b>16922</b>	<b>9693</b>	<b>57</b>	<b>751</b>	<b>37179</b>	<b>45652</b>	
5	Calcium	83	311	152	49	81	606	763
	Iron	83	444	330	74	74	925	2526
	Potassium	83	97	42	43	28	167	233
	Manganese	83	15	8	55	3	28	54
	Lead	83	15	14	91	1	43	76
	Zinc	83	110	172	156	15	415	1263
	Si	83	345	206	60	67.6	816	1139
	NO <sub>x</sub>	85	1400	900	64	100	2800	5400
	S	83	2086	1671	80	276	6175	6872
	EC	85	800	300	38	<MQL	1300	1500
	OC	85	8700	2800	32	3800	14600	17500
PM <sub>2.5</sub>	83	<b>19725</b>	<b>9150</b>	<b>46</b>	<b>5393</b>	<b>38990</b>	<b>46741</b>	
6	Calcium	142	108	67	62	<MQL	221	479
	Iron	142	153	67	44	5	264	463
	Potassium	142	60	32	53	<MQL	103	288
	Manganese	142	4	3	57	<MQL	9	15
	Lead	142	6	4	71	<MQL	12	23
	Zinc	144	28	29	105	1	79	262
	Si	142	194	192	99	<MQL	478	1714
	NO <sub>x</sub>	143	1400	1000	71	140	3600	6600
	S	142	1845	1605	87	7.8	5319	6883
	EC	147	700	300	43	100	1300	1900
	OC	147	7100	2500	35	2100	11800	16400
PM <sub>2.5</sub>	140	<b>16344</b>	<b>9040</b>	<b>55</b>	<b>4463</b>	<b>36067</b>	<b>43140</b>	
7	Calcium	94	85	65	77	22	252	412
	Iron	94	118	136	115	22	253	1283
	Potassium	94	59	37	62	22	108	296
	Manganese	94	4	6	134	1	10	56
	Lead	94	5	3	69	<MQL	10	17
	Zinc	94	22	18	83	2	52	147
	Si	94	183	251	137	<MQL	330	2158
	NO <sub>x</sub>	87	1000	800	80	200	2500	5700
	S	94	2047	1639	80	175	5522	6860
	EC	99	600	200	33	<MQL	900	1400
	OC	99	6200	2200	35	1100	10300	12500
PM <sub>2.5</sub>	92	<b>16052</b>	<b>9404</b>	<b>59</b>	<b>0</b>	<b>42467</b>	<b>39300</b>	

\*Conversions used in the manuscript to quantify SO<sub>4</sub>: SO<sub>4</sub> = S x 4.125, \*Conversions used in the Table OC = OC x 1.4

**Supplemental Table 5: Residential Outdoor Concentrations in the Enumeration Monitoring Areas (Winter)**

Enumeration Monitoring Areas	Component	N Observations	Mean (ng/m <sup>3</sup> )	Std Dev	RSD %	Minimum (ng/m <sup>3</sup> )	95 <sup>th</sup> Pctl (ng/m <sup>3</sup> )	Max (ng/m <sup>3</sup> )
1	Calcium	73	161	112	70	13	347	769
	Iron	73	325	260	80	26	893	1248
	Potassium	73	68	51	75	14	169	268
	Manganese	73	10	8	79	<MQL	22	39
	Lead	73	9	8	85	1	20	58
	Zinc	73	47	35	75	7	123	139
	Si	73	133	107	80	<MQL	272	692
	NO <sub>3</sub>	114	4500	3700	82	200	11000	16000
	S	73	876	552	63	180	2050	2433
	EC	113	600	300	50	<MQL	1100	2000
	OC	113	6500	2100	32	2700	9900	14000
PM <sub>2.5</sub>	<b>105</b>	<b>15041</b>	<b>8689</b>	<b>58</b>	<b>3231</b>	<b>32437</b>	<b>39340</b>	
3	Calcium	79	164	109	67	10	392	523
	Iron	79	236	186	79	28	674	952
	Potassium	79	66	52	79	10	187	276
	Manganese	79	9	9	92	<MQL	29	44
	Lead	79	8	7	85	<MQL	23	27
	Zinc	79	51	46	90	4	170	203
	Si	79	119	81	68	<MQL	277	431
	NO <sub>3</sub>	152	5500	5900	107	100	14300	47000
	S	79	1245	1204	97	223	4941	6414
	EC	153	600	300	50	<MQL	1200	1800
	OC	153	7300	3000	41	2000	13000	18000
PM <sub>2.5</sub>	<b>146</b>	<b>16920</b>	<b>11881</b>	<b>70</b>	<b>2431</b>	<b>34228</b>	<b>85611</b>	
4	Calcium	62	78	48	62	8	181	227
	Iron	62	110	69	63	23	268	325
	Potassium	62	51	37	73	14	100	273
	Manganese	62	4	3	68	<MQL	12	14
	Lead	62	5	4	80	<MQL	14	17
	Zinc	62	32	32	102	4	90	200
	Si	62	86	60	70	<MQL	180	520
	NO <sub>3</sub>	102	4400	3300	75	300	11000	14000
	S	62	888	488	55	145	1734	2371
	EC	105	300	200	67	<MQL	700	900
	OC	105	5900	2100	36	1600	9100	11300
PM <sub>2.5</sub>	<b>99</b>	<b>13292</b>	<b>7378</b>	<b>55</b>	<b>2016</b>	<b>29861</b>	<b>32364</b>	
5	Calcium	38	348	363	104	11	1176	1917
	Iron	38	921	1469	160	25	5530	7130
	Potassium	38	85	44	52	9	162	258
	Manganese	38	19	15	78	1	50	77
	Lead	38	42	113	273	1	240	665
	Zinc	38	92	77	84	12	319	333
	Si	38	230	178	77	<MQL	596	1003
	NO <sub>3</sub>	64	4000	3400	85	100	10000	17000
	S	38	915	424	46	212	1659	1785
	EC	65	500	700	140	<MQL	1100	2300
	OC	65	6000	2000	33	1700	9700	11000
PM <sub>2.5</sub>	<b>62</b>	<b>16573</b>	<b>6792</b>	<b>41</b>	<b>3176</b>	<b>29295</b>	<b>40687</b>	
6	Calcium	82	116	85	73	10	269	432
	Iron	82	186	138	75	23	501	714
	Potassium	82	61	52	86	4	161	385
	Manganese	82	6	5	88	0.0	17	25
	Lead	82	5	6	128	<MQL	16	34
	Zinc	82	35	37	105	2	101	192
	Si	82	105	83	79	168	224	483
	NO <sub>3</sub>	117	4800	4100	85	20	14000	25000
	S	82	1088	921	85	168	3364	4980
	EC	119	500	300	60	<MQL	900	1600
	OC	119	6300	2700	43	2000	11700	18000
PM <sub>2.5</sub>	<b>116</b>	<b>15313</b>	<b>11511</b>	<b>75</b>	<b>3176</b>	<b>40990</b>	<b>73865</b>	
7	Calcium	69	67	42	62	6	155	217
	Iron	69	87	84	97	10	229	612
	Potassium	69	50	38	75	14	116	275
	Manganese	69	4	4	97	<MQL	11	23
	Lead	69	4	5	117	<MQL	13	31
	Zinc	69	22	25	115	1	66	170
	Si	69	71	51	72	<MQL	170	193
	NO <sub>3</sub>	91	4500	4000	89	200	11000	22000
	S	69	957	775	81	216	2560	4189
	EC	95	300	300	100	<MQL	900	1200
	OC	95	5700	2700	47	50	13000	14000
PM <sub>2.5</sub>	<b>92</b>	<b>12410</b>	<b>9162</b>	<b>73</b>	<b>1667</b>	<b>33163</b>	<b>56815</b>	

\*Conversions used in the manuscript to quantify SO<sub>4</sub>: SO<sub>4</sub> = S x 4.125,

\*Conversions used in the Table OC = OC x 1.4

**Supplemental Table 6: Residential Indoor Concentrations in the Enumeration Monitoring Areas (Summer)**

Enumeration Monitoring Areas	Component	Observations	Mean (ng/m <sup>3</sup> )	Std Dev	RSD %	Minimum (ng/m <sup>3</sup> )	95 <sup>th</sup> Pctl (ng/m <sup>3</sup> )	Max (ng/m <sup>3</sup> )
1	Calcium	105	145	74	51	44	328	366
	Iron	105	254	179	70	49	643	954
	Potassium	105	51	55	108	21	218	382
	Manganese	105	8	4.5	56	0.6	15	25
	Lead	105	12	15	125	1.3	31	121
	Zinc	118	71	87	123	5	260	638
	Si	105	253	357	141	27	433	3603
	NO <sub>3</sub>	115	907	631	68	113	2108	4147
	S	105	1683	1409	84	188	4439	6474
	EC	122	859	274	32	355	1347	1548
OC	126	18313	5701	31	7802	27869	40516	
	<b>PM<sub>2.5</sub></b>	<b>105</b>	<b>19063</b>	<b>9010</b>	<b>47</b>	<b>6270</b>	<b>38148</b>	<b>44113</b>
3	Calcium	62	99	44	44	33	180	200
	Iron	62	145	103	71	20	345	513
	Potassium	62	66	35	53	8	124	175
	Manganese	62	5	3	60	1	11	13
	Lead	62	8	10	125	< MQL	21	73
	Zinc	63	38	37	97	4	131	180
	Si	62	213	215	101	30	518	1498
	NO <sub>3</sub>	64	548	442	81	37	1293	1867
	S	62	1425	1256	88	128	3903	5442
	EC	65	890	484	54	194	1860	2733
OC	65	15934	4321	27	4284	23279	29536	
	<b>PM<sub>2.5</sub></b>	<b>63</b>	<b>14949</b>	<b>8660</b>	<b>58</b>	<b>3607</b>	<b>32681</b>	<b>46422</b>
4	Calcium	125	67	31	46	22	133	184
	Iron	125	70	38	54	16	157	179
	Potassium	125	74	82	111	9	193	579
	Manganese	125	3	2	67	< MQL	7	10
	Lead	125	4	3	75	< MQL	9	22
	Zinc	125	42	106	252	3	107	858
	Si	125	203	362	178	< MQL	516	3838
	NO <sub>3</sub>	121	789	537	68	90	1672	2728
	S	125	543	1114	205	135	3587	4819
	EC	125	577	299	52	161	1154	2267
OC	125	25521	15982	63	10213	53354	143411	
	<b>PM<sub>2.5</sub></b>	<b>125</b>	<b>26884</b>	<b>26368</b>	<b>98</b>	<b>5593</b>	<b>79733</b>	<b>210724</b>
5	Calcium	80	199	121	61	43	460	774
	Iron	80	286	216	75	46	795	946
	Potassium	80	95	79	83	26	198	596
	Manganese	80	9	7	78	0.6	22	46
	Lead	80	12	10	83	< MQL	35	49
	Zinc	80	123	231	188	5	452	1774
	Si	80	247	144	58	43	553	881
	NO <sub>3</sub>	85	724	593	82	76	1843	3183
	S	80	1668	1433	86	262	5331	6496
	EC	85	820	501	61	279	1464	3482
OC	85	20521	5009	24	10891	28442	39084	
	<b>PM<sub>2.5</sub></b>	<b>80</b>	<b>20217</b>	<b>10030</b>	<b>50</b>	<b>8780</b>	<b>40120</b>	<b>55193</b>
6	Calcium	139	79	43	54	19	185	273
	Iron	139	92	50	54	10	159	294
	Potassium	139	33	60	182	8	170	411
	Manganese	139	3	2	67	< MQL	6	10
	Lead	139	4	3	75	< MQL	10	17
	Zinc	143	20	22	110	2	56	201
	Si	139	310	1254	405	< MQL	484	12553
	NO <sub>3</sub>	143	586	532	91	38	1712	2657
	S	139	1434	1289	90	149	3807	5630
	EC	136	675	561	83	< MQL	1180	6365
OC	136	20144	5494	27	4992	31096	44898	
	<b>PM<sub>2.5</sub></b>	<b>139</b>	<b>18362</b>	<b>14597</b>	<b>79</b>	<b>3087</b>	<b>38876</b>	<b>140632</b>
7	Calcium	27	54	19	35	19	88	93
	Iron	27	43	40	93	9	85	219
	Potassium	27	44	26	59	11	88	117
	Manganese	27	3	4	142	< MQL	7	13
	Lead	27	2	3	113	< MQL	6	9
	Zinc	30	11	7	66	2	23	34
	Si	27	133	121	91	18	350	612
	NO <sub>3</sub>	30	426	480	113	< MQL	1392	1826
	S	27	896	974	109	67	3395	3621
	EC	30	395	219	55	< MQL	789	911
OC	30	16392	5465	33	5768	29467	30226	
	<b>PM<sub>2.5</sub></b>	<b>27</b>	<b>11510</b>	<b>9615</b>	<b>84</b>	<b>1086</b>	<b>33482</b>	<b>35626</b>

\*Conversions used in the manuscript to quantify SO<sub>4</sub>: SO<sub>4</sub> = S x 4.125, Conversions used in the Table OC = OC x 1.4

**Supplemental Table 7: Residential Indoor Concentrations in the Enumeration Monitoring Areas (Winter)**

Enumeration Monitoring Areas	Component	Observations	Mean (ng/m <sup>3</sup> )	Std Dev	RSD %	Minimum (ng/m <sup>3</sup> )	95 <sup>th</sup> Pctl (ng/m <sup>3</sup> )	Max (ng/m <sup>3</sup> )
1	Calcium	69	269	784	291	23	755	4824
	Iron	69	173	143	83	31	449	772
	Potassium	69	89	81	91	14	304	399
	Manganese	69	5	4	80	< MQL	12	24
	Lead	69	6	5	83	1	10	40
	Zinc	69	41	48	117	2	94	312
	Si	69	289	594	206	19	1061	3314
	NO <sub>3</sub>	99	1156	1199	104	95	3788	5992
	S	69	666	500	75	126	1428	3392
	EC	99	628	1445	230	< MQL	1159	13595
	OC	99	24701	8588	35	13631	42276	69504
PM <sub>2.5</sub>	<b>99</b>	<b>16004</b>	<b>14007</b>	<b>87</b>	<b>0</b>	<b>39585</b>	<b>95517</b>	
3	Calcium	72	74	41	55	15	157	227
	Iron	72	118	113	96	14	458	512
	Potassium	72	55	47	85	6	173	236
	Manganese	72	5	6	120	< MQL	22	26
	Lead	72	5	5	100	< MQL	16	23
	Zinc	72	33	32	97	2	117	135
	Si	72	112	111	99	< MQL	277	670
	NO <sub>3</sub>	76	1089	1335	122	< MQL	3750	7029
	S	72	885	890	100	112	3354	3644
	EC	77	500	374	75	< MQL	1157	1823
	OC	77	18948	4520	24	2402	25325	30547
PM <sub>2.5</sub>	<b>70</b>	<b>10462</b>	<b>7471</b>	<b>71</b>	<b>0</b>	<b>26938</b>	<b>33404</b>	
4	Calcium	55	61	49	80	14	168	284
	Iron	55	59	40	68	11	148	179
	Potassium	55	103	170	165	11	588	814
	Manganese	55	3	2	67	0	7	7.6
	Lead	55	3	3	100	< MQL	10	16
	Zinc	55	22	23	104	3	70	138
	Si	55	100	121	121	< MQL	350	604
	NO <sub>3</sub>	66	907	892	98	18	2817	4162
	S	55	598	332	56	69	1320	1436
	EC	74	494	1680	340	< MQL	963	14587
	OC	74	32714	21052	64	13229	72750	139060
PM <sub>2.5</sub>	<b>74</b>	<b>26577</b>	<b>34411</b>	<b>129</b>	<b>1932</b>	<b>96342</b>	<b>200786</b>	
5	Calcium	29	153	125	82	14	378	774
	Iron	29	252	232	92	39	439	617
	Potassium	29	80	50	63	25	182	230
	Manganese	29	8	5	63	0.5	21	22
	Lead	29	14	37	264	01	22	665
	Zinc	29	60	56	93	7	148	286
	Si	29	219	182	83	42	552	821
	NO <sub>3</sub>	52	1593	2333	146	< MQL	6750	12339
	S	29	667	285	43	219	1114	1234
	EC	55	503	778	155	< MQL	2983	3803
	OC	55	27475	9263	34	15213	47537	67292
PM <sub>2.5</sub>	<b>54</b>	<b>21042</b>	<b>16974</b>	<b>81</b>	<b>2713</b>	<b>60251</b>	<b>85782</b>	
6	Calcium	81	60	43	72	15	127	284
	Iron	81	69	49	71	6	146	344
	Potassium	81	46	35	76	7	118	192
	Manganese	81	2	2	100	< MQL	6	12
	Lead	81	2.9	3.4	117	< MQL	10	17
	Zinc	81	18	17	94	1	50	97
	Si	81	86	83	97	< MQL	274	413
	NO <sub>3</sub>	97	789	769	97	45	2256	5105
	S	81	581	392	67	125	1322	2136
	EC	103	377	328	87	< MQL	969	1867
	OC	103	26504	19622	74	1985	45266	17827
PM <sub>2.5</sub>	<b>102</b>	<b>16599</b>	<b>31433</b>	<b>189</b>	<b>1101</b>	<b>44146</b>	<b>297980</b>	
7	Calcium	35	52	27	52	15	114	149
	Iron	35	53	47	59	8	167	221
	Potassium	35	40	25	63	12	91	131
	Manganese	35	3	2	67	0	7	9
	Lead	35	3	4	133	< MQL	12	14
	Zinc	35	16	15	94	2	54	69
	Si	35	68	61	90	< MQL	159	313
	NO <sub>3</sub>	32	527	600	114	< MQL	2125	2143
	S	35	647	552	85	113	1920	2012
	EC	35	455	231	51	116	1021	1036
	OC	35	20759	6008	29	7016	33675	36555
PM <sub>2.5</sub>	<b>35</b>	<b>9652</b>	<b>8937</b>	<b>93</b>	<b>1050</b>	<b>29448</b>	<b>40748</b>	

\*Conversions used in the manuscript to quantify SO<sub>4</sub>: SO<sub>4</sub> = S x 4.125,

\*Conversions used in the Table OC = OC x 1.4

**Supplemental Table 8: Personal Exposures in the Enumeration Monitoring Areas (Summer)**

Enumeration Monitoring Areas	Component	Observations	Mean (ng/m <sup>3</sup> )	Std Dev	RSD %	Minimum (ng/m <sup>3</sup> )	95 <sup>th</sup> Petl (ng/m <sup>3</sup> )	Max (ng/m <sup>3</sup> )
1	Calcium	107	158	117	74	0	278	1131
	Iron	107	239	169	71	<MQL	560	821
	Potassium	71	105	74	71	4	227	391
	Manganese	107	8	5	65	1	20	22
	Lead	71	11	16	144	<MQL	45	93
	Zinc	112	67	80	119	1	235	509
	Si	107	283	266	94	<MQL	832	1507
	S	122	1997	1422	82	<MQL	5501	6986
PM <sub>2.5</sub>	<b>106</b>	<b>19535</b>	<b>9805</b>	<b>50</b>	<b>4769</b>	<b>42082</b>	<b>49313</b>	
3	Calcium	61	138	93	68	26	304	521
	Iron	61	171	142	83	19	379	952
	Potassium	61	85	56	67	<MQL	198	266
	Manganese	61	7	5	83	<MQL	18	28
	Lead	61	8	11	129	<MQL	18	60
	Zinc	64	44	66	149	2	123	507
	Si	61	285	278	98	<MQL	833	1556
	S	61	1390	1222	88	33	3978	5524
PM <sub>2.5</sub>	<b>59</b>	<b>17761</b>	<b>8755</b>	<b>49</b>	<b>7014</b>	<b>31611</b>	<b>51858</b>	
4	Calcium	107	104	201	192	12	192	1757
	Iron	107	97	101	105	13	219	797
	Potassium	67	71	75	106	<MQL	166	556
	Manganese	108	4	4	100	<MQL	10	31
	Lead	67	4	5	124	<MQL	11	23
	Zinc	109	48	155	323	<MQL	100	1173
	Si	107	250	465	186	<MQL	718	4126
	S	107	1389	1068	77	113	3668	4779
PM <sub>2.5</sub>	<b>108</b>	<b>26238</b>	<b>24588</b>	<b>94</b>	<b>1030</b>	<b>64684</b>	<b>179013</b>	
5	Calcium	75	203	119	59	39	525	721
	Iron	75	263	186	71	28	736	843
	Potassium	36	109	104	96	<MQL	315	611
	Manganese	75	8	5	61	<MQL	19	24
	Lead	36	11	7	66	<MQL	22	39
	Zinc	75	101	168	166	3	356	1199
	Si	75	296	200	68	<MQL	678	1010
	S	75	1591	1479	93	134	5355	6504
PM <sub>2.5</sub>	<b>76</b>	<b>23820</b>	<b>16335</b>	<b>69</b>	<b>3241</b>	<b>53781</b>	<b>111383</b>	
6	Calcium	125	92	67	72	26	244	409
	Iron	125	130	223	172	18	236	2289
	Potassium	78	70	57	83	1	191	289
	Manganese	125	5	9	196	<MQL	10	90
	Lead	78	5	5	100	<MQL	15	27
	Zinc	129	30	55	186	2	89	566
	Si	125	213	206	97	<MQL	661	1232
	S	125	1426	1283	90	126	3938	5574
PM <sub>2.5</sub>	<b>124</b>	<b>18779</b>	<b>10843</b>	<b>58</b>	<b>3497</b>	<b>38820</b>	<b>81436</b>	
7	Calcium	29	98	82	84	15	340	346
	Iron	29	69	56	81	17	206	250
	Potassium	29	56	40	72	<MQL	139	141
	Manganese	29	3	3	100	<MQL	8	8
	Lead	29	4	7	186	<MQL	11	35
	Zinc	30	28	64	228	1	64	357
	Si	29	359	443	123	<MQL	1052	2248
	S	29	955	963	101	96	3240	3556
PM <sub>2.5</sub>	<b>29</b>	<b>13853</b>	<b>9837</b>	<b>71</b>	<b>1850</b>	<b>34427</b>	<b>39305</b>	

<sup>a</sup> Conversions used in the manuscript to quantify SO<sub>4</sub>: SO<sub>4</sub> = S x 4.125

<sup>b</sup> EC/ OC not taken for personal

**Supplemental Table 9: Personal Exposures in the Enumeration Monitoring Areas (Winter)**

Enumeration Monitoring Areas	Component	Observations	Mean (ng/m <sup>3</sup> )	Std Dev	RSD %	Minimum (ng/m <sup>3</sup> )	95 <sup>th</sup> Pctl (ng/m <sup>3</sup> )	Max (ng/m <sup>3</sup> )
1	Calcium	60	206	477	232	28	1035	2585
	Iron	60	186	218	117	18	541	1520
	Potassium	60	83	79	95	2	222	445
	Manganese	60	7	9	132	<MQL	15	62
	Lead	60	6	6	100	<MQL	13	32
	Zinc	60	50	91	181	4	151	651
	Si	60	238	383	161	< MQL	1052	1993
	S	60	603	404	67	40	1474	1941
	<b>PM<sub>2.5</sub></b>	<b>67</b>	<b>16192</b>	<b>11942</b>	<b>74</b>	<b>1566</b>	<b>37955</b>	<b>68659</b>
3	Calcium	72	101	90	90	24	211	640
	Iron	72	126	122	97	14	466	556
	Potassium	72	63	53	83	<MQL	170	265
	Manganese	72	5	6	108	<MQL	18	28
	Lead	72	6	7	119	<MQL	22	28
	Zinc	72	33	33	100	2	124	150
	Si	72	151	138	91	< MQL	473	608
	S	72	823	887	108	76	3322	4021
	<b>PM<sub>2.5</sub></b>	<b>72</b>	<b>12946</b>	<b>10563</b>	<b>82</b>	<b>803</b>	<b>32017</b>	<b>65618</b>
4	Calcium	49	67	53	80	17	156	357
	Iron	49	65	41	63	9	138	179
	Potassium	49	126	220	175	<MQL	642	1049
	Manganese	49	3	2	75	<MQL	8	10
	Lead	49	3	4	135	<MQL	10	14
	Zinc	49	24	23	93	2	73	112
	Si	49	134	151	113	< MQL	568	679
	S	49	586	315	54	84	1235	1310
	<b>PM<sub>2.5</sub></b>	<b>69</b>	<b>33348</b>	<b>41970</b>	<b>126</b>	<b>2184</b>	<b>113585</b>	<b>225361</b>
5	Calcium	28	134	99	74	42	290	522
	Iron	28	232	192	83	24	426	1011
	Potassium	28	76	48	63	8	175	191
	Manganese	28	8	5	67	<MQL	18	24
	Lead	28	13	35	270	<MQL	23	186
	Zinc	28	49	36	73	7	117	170
	Si	28	204	152	75	< MQL	501	670
	S	28	659	300	46	194	1149	1186
	<b>PM<sub>2.5</sub></b>	<b>46</b>	<b>23514</b>	<b>21852</b>	<b>93</b>	<b>1339</b>	<b>50479</b>	<b>129312</b>
6	Calcium	69	75	56	74	3	189	353
	Iron	69	77	76	98	5	206	509
	Potassium	69	49	39	80	<MQL	126	252
	Manganese	69	3	3	100	<MQL	8	12
	Lead	69	3	5	167	<MQL	11	15
	Zinc	69	25	31	124	1	74	202
	Si	69	113	121	107	< MQL	351	601
	S	69	541	419	77	7.2	1402	1990
	<b>PM<sub>2.5</sub></b>	<b>89</b>	<b>17887</b>	<b>29819</b>	<b>167</b>	<b>&lt; MQL</b>	<b>54600</b>	<b>255837</b>
7	Calcium	29	119	152	127	22	284	815
	Iron	29	67	72	107	11	187	347
	Potassium	29	52	27	52	12	107	121
	Manganese	29	4	3	75	<MQL	10	14
	Lead	29	3	6	200	<MQL	15	20
	Zinc	29	27	31	114	1	88	154
	Si	29	209	259	124	< MQL	834	1057
	S	29	670	628	94	112	2063	2539
	<b>PM<sub>2.5</sub></b>	<b>29</b>	<b>10351</b>	<b>6769</b>	<b>65</b>	<b>2137</b>	<b>24082</b>	<b>24581</b>

<sup>a</sup> Conversions used in the manuscript to quantify SO<sub>4</sub>: SO<sub>4</sub> = S x 4.125

<sup>b</sup> EC/ OC not taken for personal

**Supplemental Table 10: Indoor vs. Outdoor Ratios**

<b>Component</b>	<b>Season</b>	<b>Observations</b>	<b>Factor</b>
<b>Calcium</b>	<b>Summer</b>	521	0.7
	<b>Winter</b>	332	1.4
<b>Iron</b>	<b>Summer</b>	521	0.7
	<b>Winter</b>	332	0.5
<b>Potassium</b>	<b>Summer</b>	521	1.2
	<b>Winter</b>	332	1.4
<b>Manganese</b>	<b>Summer</b>	521	0.8
	<b>Winter</b>	332	0.5
<b>Lead</b>	<b>Summer</b>	521	0.7
	<b>Winter</b>	332	0.2
<b>Zinc</b>	<b>Summer</b>	546	1.6
	<b>Winter</b>	332	0.8
<b>Silicon</b>	<b>Summer</b>	521	1.4
	<b>Winter</b>	332	1.3
<b>Nitrates</b>	<b>Summer</b>	557	0.6
	<b>Winter</b>	322	0.3
<b>Sulfur</b>	<b>Summer</b>	521	1.0
	<b>Winter</b>	332	0.7
<b>PM<sub>2.5</sub></b>	<b>Summer</b>	560	1.0
	<b>Winter</b>	344	1.0

Supplemental table 11:

## Personal vs. Indoor Ratios

Component	Season	Observations	Factor
Calcium	Summer	479	1.4
	Winter	302	1.0
Iron	Summer	479	1.4
	Winter	302	1.0
Potassium	Summer	321	1.2
	Winter	302	1.0
Manganese	Summer	521	1.4
	Winter	332	1.9
Lead	Summer	321	1.1
	Winter	302	0.9
Zinc	Summer	509	1.6
	Winter	302	1.0
Silicon	Summer	479	1.2
	Winter	302	1.1
Sulfur	Summer	479	1.0
	Winter	302	1.0
PM <sub>2.5</sub>	Summer	477	1.2
	Winter	299	1.1

**Supplemental Table 12: MIXED Model Results – Examining the effect of spatial variability using average mean concentrations (ng/m<sup>3</sup>) of personal measures adjusting for ambient air by season.**

Component	Season	Enumeration Monitoring Areas (EMA)						p-Value
		1	3	4	5	6	7	
Calcium	Summer	159	141	111	200	97	103	< 0.01
	Winter	199	103	66	128	74	119	0.178
Iron	Summer	246	167	99	262	134	73	< 0.01
	Winter	192	120	68	233	77	61	< 0.01
Potassium	Summer	104	87	75	120	71	56	0.040
	Winter	88	60	153	76	48	50	0.123
Manganese	Summer	8	7	4	8	5	3	< 0.01
	Winter	7	5	3	8	3	3	< 0.01
Lead	Summer	11	8	4	11	5	4	< 0.01
	Winter	6	6	3	13	4	3	< 0.01
Zinc	Summer	66	47	55	94	29	33	0.12
	Winter	51	31	26	51	25	24	0.02
Silicon	Summer	289	289	209	304	222	361	0.08
	Winter	240	151	132	205	111	208	0.03
Sulfur	Summer	1596	1435	1197	1586	1396	899	< 0.01
	Winter	742	707	638	743	535	523	0.02
PM <sub>2.5</sub>	Summer	20,400	17,800	24,900	24,100	19,000	13,600	0.031
	Winter	16,200	12,400	34,300	26,900	16,000	9,700	0.014